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Seawater desalination: nanofiltration—a substitute for reverse osmosis?

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ABSTRACT

We present the possibility to replace the first-stage reverse osmosis (RO) seawater by nanofiltration (NF) membranes. Then, we tested few membranes in the laboratory (NF200, NF2540, MPS44, and MPS34) on synthetic and natural seawater, for 2.6 m^2 membrane area pilot. We observed that the membrane NF200 is the best membrane from all tested due to its higher productivity with $1.8\,L\,h^{-1}\,m^{-2}\,bar^{-1}$ and its higher salt rejection with 60%. Furthermore, as expected, NF200 filtration of natural seawater permits to decrease SDI_3 to a value of $4 \pm 2\%$/min and total organic carbon under 0.1 mg/L, eliminating totally the risk of biofouling development on second-stage RO treatment. The configuration NF200-SW30 gave the best energy gain with 29%. The permeate obtained presents a total dissolved solids of 324 ppm and the flow yield ratio increase of 16%, in reference to SW30-SW30 configuration. Also, we tested the limits of ROSA^{TM} simulation, especially for NF membranes. We observed lots of flaws, especially for high pressures and high salts concentrations. As a consequence, each simulation realized for NF must be validated by experiments on real seawater to a sufficient scale and during sufficient time of experiments.

Keywords: Membrane; Reverse osmosis; Nanofiltration; Mass transfer modeling; Simulation; Pretreatment; Seawater

1. Introduction

Water is necessary to all human activities on Earth. Resources decrease in some places of the globe and the significant increase of human population intensifies the need for water and the availability issue. Two new technologies are implemented to produce water for human consumption from seawater: thermal and reverse osmosis (RO) membrane processes.

RO technique is the most used (Fig. 1). But the main issue with RO is its great sensitivity to (bio)fouling that impacts on the membrane lifetime and therefore on the process costs [1]. The present work gathers first a comprehensive bibliographic review on the positioning of NF as pretreatment prior to RO and describes in detail the initial work done in this field relating to the creation of the first plant in the world, Umm Lujj in Saudi Arabia [2]. We will also detail the experience of seawater NF for the preparation of isotonic/hypertonic waters for human diet [3,4]. NF was also very well developed in few fields of application, as reported by Schäfer et al. [5].

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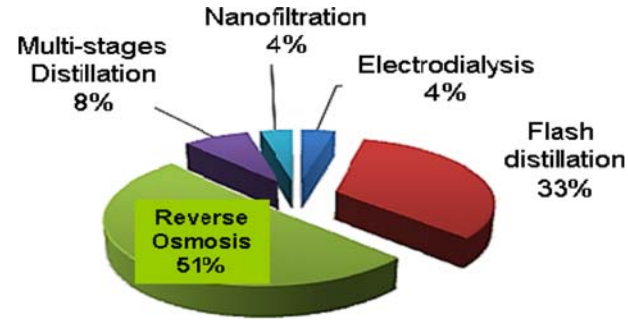


Fig. 1. Installed plant in the world for water desalination technics.

The work will report also seawater NF to replace RO seawater at its first stage. To validate such research, we conducted simulation with ROSA™ software and the predictions obtained were compared to experimental results on artificial (NaCl solution of 35 g/L) and real seawater collected at Le Croisic (Loire-Atlantique, west of France).

2. Background

Twenty years ago, the high price of desalting membranes has limited their applications to very low brackish waters (under 5 g/L of total salinity) and limited their development to small filtration plants, with low-pressure RO, that is, the membrane BW30, purchased by Dow (US) company. Today, the ability of novel NF membranes for a more selective demineralization of salty solutions using low costs materials makes it an interesting technique in many respects [6–10]. Then, we have decided to compare an old generation of thin film composite NF membrane (denoted NF70) with a novel generation of sulphonated (–SO_3^- polypyperazine amide membrane (denoted NF200) developed especially to decrease by half the well-known softening properties of NF for the elaboration of drinking water from the river Oise, (West of Paris, France) [11]. We proposed presently to study the membrane NF200 as seawater RO pretreatment.

Previous studies conducted in the beginning of the twenty-first century compared NF membranes with RO for seawater pretreatment to replace the usual first pass of RO [2]. Furthermore, the authors have reported the interest of NF as a selective demineralization between monovalents/divalent ions, that is, Na^+, Ca^{++}, and Mg^{++} separation [10].

3. Material and methods

3.1. Simulation parameters

We used two versions of the software ROSA^{TM}, 6.0 and 7.2. This simulator is based on a model of mass transfer denoted solubilization–diffusion, ideal for dense RO membranes.

The operating parameters of the simulations conducted are reported in the Tables 1 and 2.

3.2. Seawater quality

First, we used a NaCl solution at 35 g/L and we extended the simulation to real seawater with the average Atlantic seawater, as detailed in the Table 3.

3.3. Pilot design

The RO/NF laboratory pilot elements are described in the Fig. 2.

The transmembrane pressure changed between 0 to 30 ± 1 bar. A dozen membranes were disposable in our laboratory. We tested all those membranes and reported presently only the acceptable results (see Fig. 3).

Table 1
Operating parameters of the simulator ROSA^{TM} in the case of modules of size 25–40 (2.6 m^2 of membrane geometrical area)

Parameters	Signs	Values	Units
Flow rate of feed	Q_0	0.6	$m^3\,h^{-1}$
Concentration in NaCl	[NaCl]	35.000	$mg\,L^{-1}$
Transmembrane pressure applied	ΔP	1–35	bars
Temperature	T	20	°C
pH	pH	7.6	/
Pump efficiency	/	80	%
Number of stages	/	1	/
Number of module by tube	/	1	/
Number of tube per stage	/	1	/
Module type	/	2,540 (membrane area 2.6 m^2)	/

Table 2
Operating parameters for pilot scale simulations in reference to Umm Lujj plant

	NF90	NF200	XLE	SW30	NF Umm Lujj	RO Umm Lujj
First stage 1 (E1) per pressure vessel						
Q_0 (m^3/h)	13.33	13.33	13.33	13.33	13.33	5.22
P (bar)	35	35	35	65	25	65
TDS feed (ppm)	35.112	35.112	35.112	35.112	45.460	45.460
Q_P (m^3/h)	4.29	6.72	3.52	6.37	8.67	1.57
Y (%)	32	50	26	48	65	30
	SW30	SW30	SW30	SW30	RO Umm Lujj	RO Umm Lujj
Second stage 2 (E2) per pressure vessel						
Q_0 (m^3/h)	4.29	6.72	3.52	6.37	19.50	7.71
P (bar)	10	30	8	5,2	64	30
TDS feed (ppm) = TDS perm. E1	4,140	13,098	2,217	426	28,260	–
Q_P (m^3/h)	2.09	3.97	2.11	1.85	11.3	6.6
Y (%)	49	59	60	29	58	85

Table 3
Seawater mineral composition of Atlantic Ocean

Cations	Concentrations (ppm)	Anions	Concentrations (ppm)
Sodium Na^+	10,770	Chlorides Cl^-	19374.14
Magnesium Mg^{2+}	1,290	Sulfates SO_4^{2-}	2,712
Calcium Ca^{2+}	412.1	Bicarbonates HCO_3^-	140
Potassium K^+	399	Carbonates CO_3^{2-}	5.884
Strontium Sr^{2+}	7.9	Fluorides F^-	1.3
TDS = 35,112.3 ppm			

We maintained all membranes in a solution of sodium bisulfite at 1% in order to prevent them from biofilm growth The Table 4 presents the studied membranes whose results are presently reported.

Before experiments, the membranes were cleaned by means of standard procedures to remove preservatives and rinsed with UP MilliQ water (Millipore, France) until the conductivity of the permeate remained below 1 μS/cm. The temperature was maintained at 20°C.

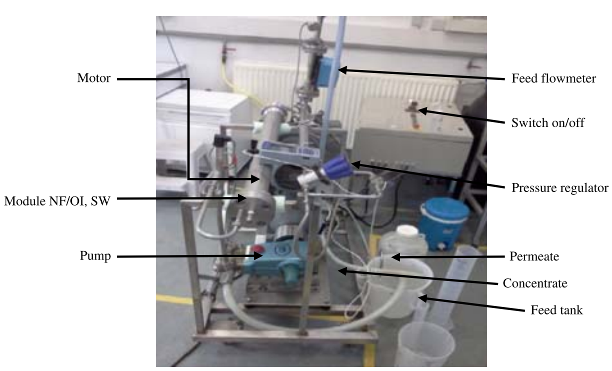


Fig. 2. Pilot for laboratory scale experiments equipped with membrane module 25–40.

3.4. Seawater sampling

Seawater was sampled in the port of «Le *Croisic* », a little town in Loire-Atlantique (west of France, in the French Brittany), as illustrated in the Fig. 4.


Fig. 3. Membranes modules 2,540 stocked in 1% sodium bisulfite solution (photography J.-S. Derauw, 05/04/2011).

Table 4
Characteristics of the tested membranes

Membrane type	Company	Type	S (m^2)	Product name	Serial number
SW30	Dow Filmtec	OI	2.8	SW30-2540-F	A8418718
NF200	Dow Filmtec	NF	2.6	NF200-2540	F3996982
NF2540	Dow Filmtec	NF	2.6	NF2540	–
XLE	Dow Filmtec	OI	2.6	–	Simulation only
MPS44	Kiryat Weizmann Ltd.	NF	1.6	MPS-44-2540-B2-X	1,656
MPS34	Koch	NF	1.6	MPS-34-2540-B2-X	–

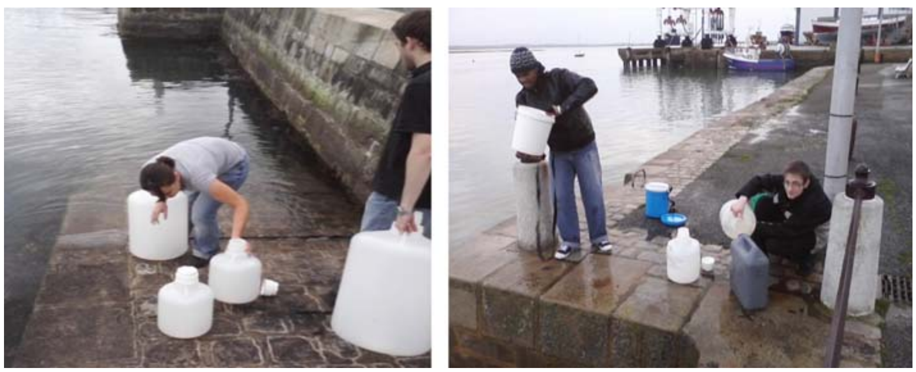


Fig. 4. Photographies of seawater sampling in Le Croisic (France).

GPS coordinates of sampling were:

- Latitude: 47° 17´ 38,09´´ Nord
- Longitude: 2° 30′32,07´´ Ouest

Days and hour of sampling: 5 June 2011 at 10 h am, 4 January 2012 at 10 h am, and 26 January 2012 at 9 h am.

4. Results and discussion

4.1. Preliminary results

In this first part, we reported some previous results [3,4] obtained in our group on seawater, comparing NF70 and NF200 membrane, at a pilot scale (membrane area 2.6 m^2) to propose a one-step operation under 25 bars for the elaboration of personal body washing solutions (salinity near 9 g/L) and dedicated to thalassotherapy.

In the same study, we prepared also very high concentrated solutions (total salinity around 70 g/L) with a high concentration of Ca^{2+}, Mg^{2+}, Zn^{2+}, and others biologically active ions for thalassotherapy centers or also for home bath washing. Then, the possibilities of NF reducing both the overall salinity of natural salting waters lead the way for potential applications in the field of human health (i.e. preparation of nasal sprays, medical dietetics, and hot mineral springs).

In 2002, our studies have extended the range of NF applications to seawater with a permeate flux obtained five times higher in NF than RO with a critical pressure attained (10 bars) three times lower than the theoretical osmotic pressure of 28 bars (see Fig. 5).

Our main objective was to validate NF operations in two of more filtration stages for the preparation of isotonic solutions (total salinity around 9 g/L).

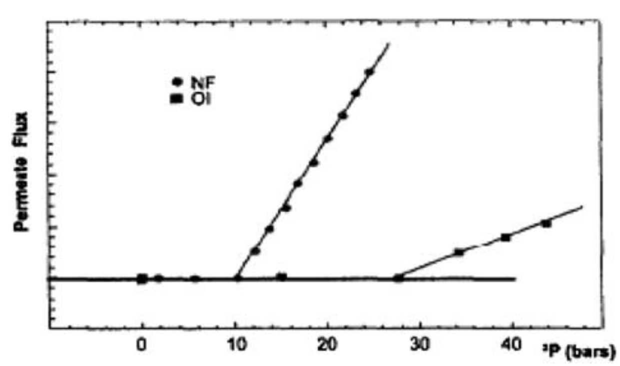


Fig. 5. Comparison of NF and RO permeability to seawater (from Pontié et al. [3]) NF90 and ESPA3 membranes.

Table 5
Permeate and concentrate solutions compositions elaborated from seawater with NF70 and NF200 membranes (from [4])

	Seawater	Isotonic permeate after 1 stage NF70 (25 bar)	Isotonic permeate after 4 stage NF200 (each step 10 bar)	NF70 concentrate
Total salinity, g/L	35	9	9	70
Na^+, mg/L	10,490	3,980	4,012	19,870
Ca^{2+}, mg/L	407	14	26	1,090
Mg^{2+}, mg/L	1,075	26	4	ND*

*ND = not determined.

The results reported in Table 5 have confirmed the very open possibilities of NF to elaborate isotonic water from seawater. Using NF70 membrane, the operation occurred only in one-stage filtration for a transmembrane pressure of 25 bar with 2 m^2 membrane area. NF200 membrane has shown a four-stage filtration under 10 bars, each stage to reach the same objective. We reported also in the Table 5 the compositions of the permeates obtained at 9 g/L and the concentrated solutions of the first NF70 stage in terms of total salinity, sodium, calcium, and magnesium concentrations.

Further results illustrating the selectivity observed in NF70 membranes with the ions Mg^{2+}, Ca^{2+} and Na^+. The rejection order followed very well the Hofmeister serie of the hydration energy, that is, $Mg^{2+} > Ca^{2+} > Na^+$. It means that NF70 predominant mass transfer is a solution diffusion, like RO (see Fig. 6).

On the contrary, the selectivity between Ca^{2+} and Mg^{2+} has been inversed, as reported by [11] on surface water treatment, with the NF200 membrane. The authors have attributed the higher affinity of the NF200 membrane for calcium to the presence of sulphonated functions grafted on the membrane surface. We observed the same inversion with the same membrane on seawater [4].

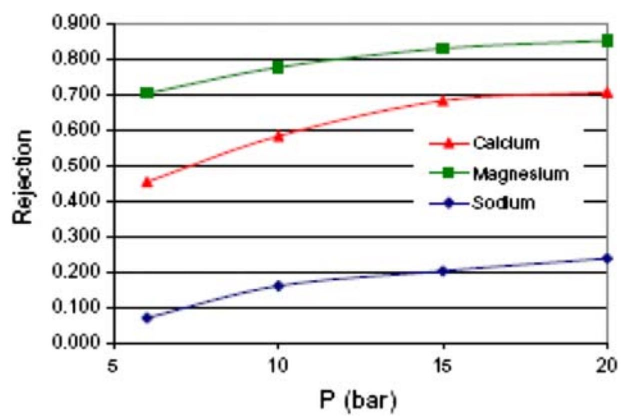


Fig. 6. Monovalent/divalent ions selectivity in NF70 membrane seawater (from Pontié et al. [4]).

4.2. Choice of the best NF membrane for seawater

The Fig. 7 (left) reports the permeate flow evolution vs. pressure, as the Fig. 7 (right) reports the retention vs. transmembrane pressure, for the four studied membrane, with a synthetic solution of NaCl at a concentration of 35 g/L.

MPS44 and MPS34 show the lowest permeabilities (Lp′) with the values of 0.53 and 1.0 L h^{-1} m^{-2} bar^{-1}, respectively. NF200 and NF2540 show the higher ones

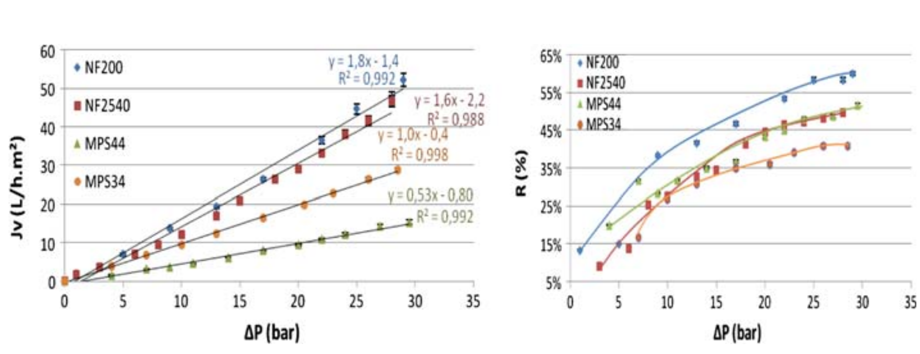


Fig. 7. $J_V = f(\Delta P)$ and $R = f$(DP) for the studied membranes (NaCl 35 g/L, feed flow 600 L/h, temperature 20°C, flow yield ratio 1–10%, module 2,540 (membrane area 2.6 m^2).

Table 6
Hydraulic permeabilities (Lp′) and salt rejection (*R*) at 30 bars for the tested membranes (NaCl solution of 35 g/L)

	NF200	NF2540	MPS44	MPS34	SW30
Lp′ ($L\,h^{-1}\,m^{-2}\,bar^{-1}$)	1.8 ± 0.2	1.6 ± 0.2	0.53 ± 0.05	1.0 ± 0.1	0.67 ± 0.05
R (%) (30 bar)	60	52	52	40	98

with 1.8 and 1.6 $L\,h^{-1}\,m^{-2}\,bar^{-1}$, respectively, as reported in the Table 6.

Retention of salts is also an essential parameter to discriminate membranes. As expected, the SW30 presents the higher retention with a value at 98% for a pressure of 30 bars. The membrane NF200 shows a value at 60% and the others membranes have lower salt retention.

To conclude, the NF200 after the essays with the membrane in our laboratory is a good candidate to replace SW30.

4.3. NF200 tests in first-stage desalination of real seawater

We tested the membrane NF200 in a first stage of seawater desalination, as RO pretreatment, modelizing the first module of a tube of six modules (see Fig. 8).

We determined the following parameters:

- Silt density index (SDI).
- Total organic carbon (TOC).
- Turbidity.

as reported in the Table 7. We pre-filtered seawater on 1 micrometer membrane nominal pore size, before nanofiltration (NF) operation.

SDI_{15} was not measurable on natural seawater. Then, we determined the SDI_3, as usually preconized [8]. Initially, we obtained a value of 19 ± 2%/min on direct seawater, value in the range of others natural seawater in the world with SDI_3 reported between 5 and 35%/min. Furthermore, pre-filtration to 1 micrometer has no influence on the fouling properties of seawater, SDI stays constant, but the turbidity in the same time decreases from 9.7 to 2.2. Maybe larger particles than SDI membrane pore size (0.45 m) are retained but without influencing SDI values. NF200 filtration of seawater permits to decrease the SDI_3 to a value of 4 ± 2%/min. Furthermore, due to the very low TOC in the NF200 permeate (<0.1 mg/L), the biofouling development on the RO membrane of the second pass should be very limited. But, as a consequence, the durability of NF membranes will depend on the pretreatments developed before, as also encountered in Mery/oise [11] where the main problem is the biofouling due to total retention of BDOC (biodegradable dissolved organic carbon) by the membrane NF200 itself.

Fig. 8. Head module of a tube (module NF200 2540).

4.3. Limits of ROSA^{TM} model in NF

The present study focused on the assessing of NF performance as a pretreatment for seawater before RO, for increasing the lifetime of RO membranes by decreasing transmembrane pressure and thereby the cost of desalination operation. NF of seawater is then examined vs. RO, both through laboratory experiments and assisted by the simulator ROSA™. But as well known, ROSA™ software is based on a solution diffusion model of mass transfer and this mechanism is predominant in RO only, but not in NF where the active layer present some microporosities. We have

Table 7
Measurements of SDI, TOC, and Turbidity on seawater and after NF

	Seawater without pre-filtration	Seawater pre-filtered on 1 μm	Permeate NF
SDI_3 (%/min) ± 2	19	21	4
SDI_5 (%/min) ± 2	Not determined	Not determined	3
TOC (ppm)	1.12 ± 0.04	0.81 ± 0.03	<0.1
Turbidity (NTU) ± 0.1	9.7	2.2	<0.5

not considered charges interactions because in the presence of seawater, as well known, all the electrostatic forces are screened.

We observed a hydraulic permeability higher experimentally with $1.9 \pm 0.1\,\mathrm{L\,h^{-1}\,m^{-2}\,bar^{-1}}$ as the simulation gives a value of $1.1 \pm 0.1\,\mathrm{L\,h^{-1}\,m^{-2}\,bar^{-1}}$. At the zero permeate flux J_v, we determined the critical pressure of the different membranes studied, as reported in a recent article [10]. In the case of SW30, the critical pressure is the theoretical osmotic pressure of seawater (around 25 bars), and as for the simulation and experiments of the NF200 membrane, this value changes from 9 to 7 bars, respectively.

The major explanations of the differences observed on the Fig. 9 are:

- In the case of RO vs. NF, the presence of microporosity in NF explains well the higher productivity in NF/RO.
- In comparison between simulation and experimentation for the NF200 membrane, the main explanation is based on the limit of ROSA's model to dense membrane, based on a pure solution diffusion mass transfer.

As observed in the Fig. 10, after 10 bars, the differences between experiments and simulations increase dramatically. At 30 bars, total dissolved solids (TDS) is $10\,\mathrm{g\,L^{-1}}$ in simulation and $19\,\mathrm{g\,L^{-1}}$ from the experiments. If the simulator ROSA^{TM} is very efficient for big plants design in RO, in NF, a lot of defaults are well observed, especially for high pressures and high salts concentrations in solution.

As previously reported [10], the mass transfer model SKK (Spiegler Kedem Katchalsky) is very

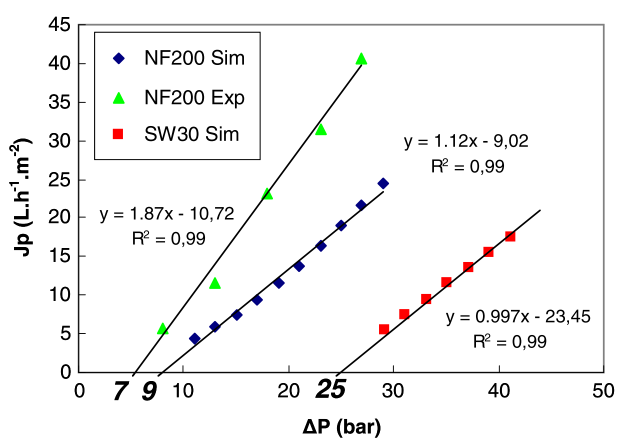


Fig. 9. Permeate flow vs. transmembrane pressure for NF200 experiments with natural seawater and simulation in comparison with simulation of the SW30.

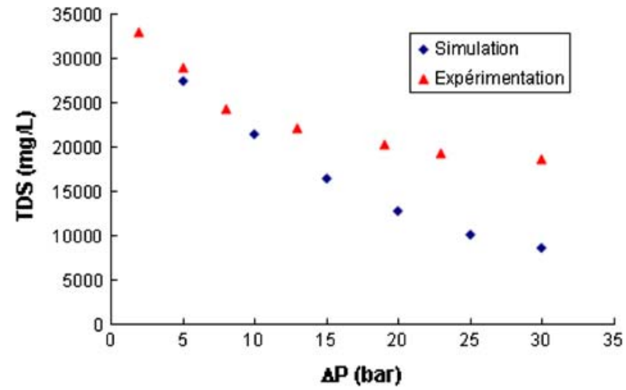


Fig. 10. TDS = f (ΔP) in a 1 stage NF200.

well adapted to explain different behavior between NF and RO. A solute mass transfer in NF and RO operations can be described by the Eq. (1), based on the following hypothesis: no polarization concentration occurred, no charge on the membrane surface, and also no variations of the permeate concentration vs. time (steady-state flow well established):

$$J_s = J_p C_p = J_{\mathrm{Diff}} + J_p C_{\mathrm{conv}} \tag{1}$$

where J_s is the solute flow, J_p is the solvent flow, J_{Diff} the solute flow due to diffusion, and $C_{\mathrm{conv.}}$ is the solute concentration in the permeate due to convection.

The above equation can be rearranged, as recently reported [10]:

$$C_p = J_{\mathrm{Diff}} \frac{1}{J_p} + C_{\mathrm{conv}} \tag{2}$$

From plot of C_p vs. $1/J_p$, each part of solute mass transfer (solution diffusion and convection) may be obtained separately.

This model was applied recently and for the first time by Ould moahmedou et al. [10] to demonstrate that an old RO membrane used for seawater desalination acquired after two years a convective mass transfer component, illustrated by an increase in membrane permeability and a dramatic decrease in the salts retention. The same approach can be presently applied by considering that experiments NF is equivalent to an old RO membrane, as simulation results are based on a pure solution diffusion model. Then, the graph $C_p = f(1/J_p)$ can be used to discriminate simulations with experiments for the NF200 membrane.

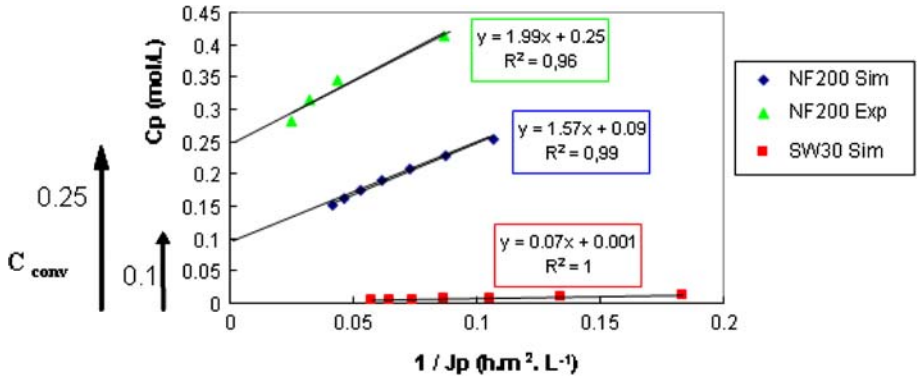


Fig. 11. Evolution de la concentration en perméat en fonction de l'inverse du flux de perméat (NF200 en simulation et expérimentation; SW30 en simulation).

Table 8
Comparison of NF200, XLE, NF90, and SW30 in terms of hydraulic permeability, critical pressure, retention of TDS, slope of $C_p = f(1/J_v)$, and C_{Conv}

Parameters/membrane	NF200 exp.	NF200 simul.	NF90 simul.	XLE simul.	SW30 simul.
Hydraulic permeability ($L\,h^{-1}\,m^{-2}\,bar^{-1}$) (±0.1)	1.9	1.1	1.0	1.6	0.9
Pc (bar) (±1)	7	9	19	22	29
Retention 35 bars (%)	35	80	90	97	99
Slope for $C_p = f(1/J_p)$ ($mol\,m^{-2}\,h^{-1}$)	1.99	1.57	0.47	0.20	0.08
C_{Conv} (g/L)	0.250	0.100	0.050	0.007	0

From the Fig. 11, the following parameters have been determined:

- hydraulic permeability, critical pressure, retention at 35 bars, J_{Diff} and C_{conv}. All those data were also determined for the membranes XLE, NF90, and SW30. All data are reported in the Table 8.

From the Table 8, we can classify the different membranes studied for the different parameters determined:

- In terms of hydraulic permeability, the following order was observed: NF200 > XLE > NF90 > SW30.
- In terms of mass transfer convection, the following order was observed: NF200 > NF90 > XLE > SW30.

The graph of the Fig. 11 is indeed very useful to classify the membranes and could facilitate the choice of the best membrane for a done application.

Also, we tested the limits of ROSA™ simulation, especially for NF membranes. A lot of defaults are well observed, especially for high pressures and high salts concentrations in solution.

Furthermore, do not forget that simulation using ROSA™ software is limited in NF. As a consequence, each simulation must be validated by experiments to do on real seawater to a sufficient scale and during sufficient time of experiments.

4.4. Simulation of different NF–RO configurations

Based on the data of the first realization in the world, pretreating seawater by NF in replacement to RO (Umm Lujj, Saudi Arabia) different configurations NF-SW30 was compared to this plant operation.

The Fig. 12 reports the flow sheet of Umm Lujj plant built in Saudi Arabia in 2002.

The design of two-stage membrane desalination unit developed in Umm Lujj is detailed in the Fig. 13. We simulated the following different NF–RO configurations: NF200-SW30, NF90-SW30, XLE-SW30 in reference to SW30-SW30. All the results obtained in terms of energy and water quality are resumed in the Fig. 14.

To do the best configuration choice, we compared the different possibilities in terms of energy gains and water quality, *in reference to SW30-SW30*. NF200-SW30

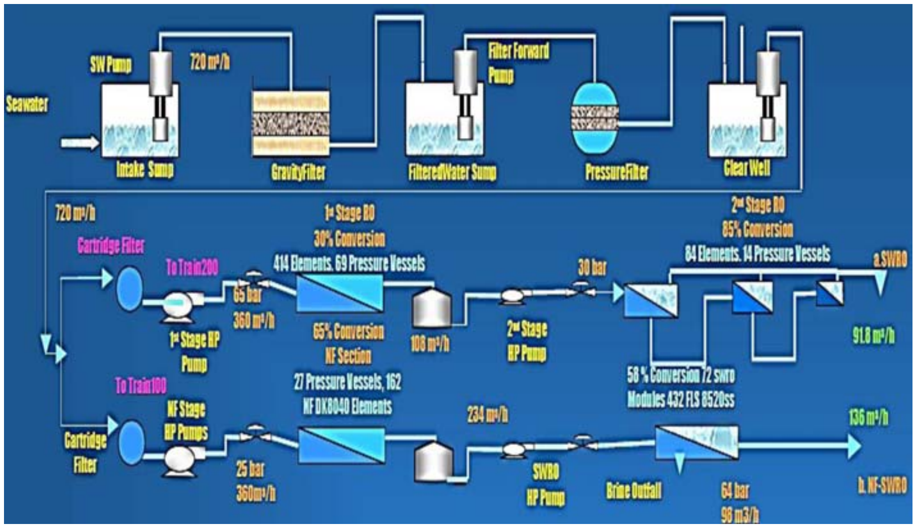


Fig. 12. Global design of Um Lujj (Saudi Arabia) realization [12].

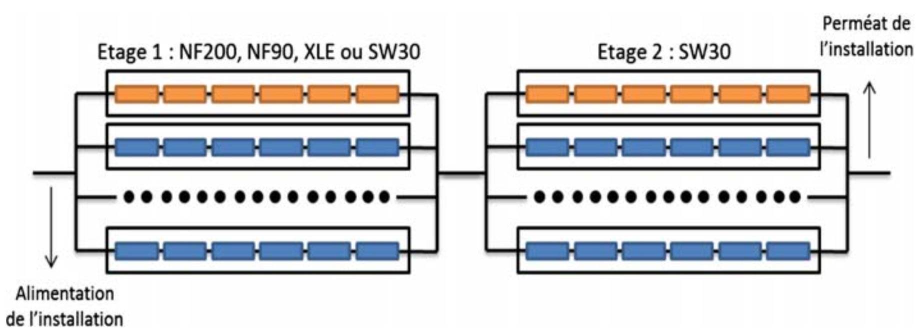
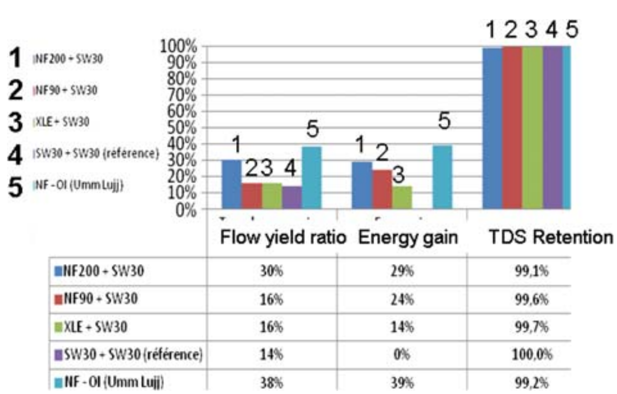


Fig. 13. Design of Umm Lujj stages 1 and 2.



	Flow yield ratio	Energy gain	TDS Retention
NF200 + SW30	30%	29%	99,1%
NF90 + SW30	16%	24%	99,6%
XLE + SW30	16%	14%	99,7%
SW30 + SW30 (référence)	14%	0%	100,0%
NF - OI (Umm Lujj)	38%	39%	99,2%

Fig. 14. Flow yield ratio, energy gains, and TDS retention for the different studied configurations.

gives the best energy gain with 29% and the permeate obtained presents a TDS of 324 ppm. Furthermore, this configuration shows a gain of 16% for the flow yield ratio. But as observed in Umm Lujj configuration NF-OI, a gain in flow yield of 8% is observed. In the case of NF200 simulation, the convective part was not taken into account and then we can project a flow yield ratio more important. We observed also that a gain in flow yield ratio correlates with a loose in water quality. The opposite result with the configurations NF90-SW30 and XLE-SW30 is observed where a gain in water quality is associated with a decrease in water productivity. So, the best balance must be optimized for each situation.

5. Conclusion

We present the possibility to replace the first-stage RO seawater by NF membranes. Then, we tested few membranes in the laboratory (NF200, NF2540, MPS44, and MPS34) on synthetic and natural seawater, for 2.6 m^2 membrane area. We observed that the membrane NF200 is the best membrane from all tested due to its higher productivity with $1.8\,L\,h^{-1}\,m^{-2}\,bar^{-1}$ and its higher salt rejection with 60%. Furthermore, as expected, NF200 filtration of natural seawater permits to decrease SDI_3 to a value of $4 \pm 2\%/min$ and TOC under 0.1 mg/L, eliminating totally the risk of biofouling development on second-stage RO treatment.

The configuration NF200-SW30 gave the best energy gain with 29%. The permeate obtained presents a TDS of 324 ppm and the flow yield ratio increase of 16%, in reference to SW30-SW30 configuration.

Also, we tested the limits of ROSA™ simulation, especially for NF membranes. We observed lots of flaws, especially for high pressures and high salts concentrations. As a consequence, each simulation realized for NF must be validated by experiments on real seawater to a sufficient scale and during sufficient time of experiments.

As a perspective, others configurations must be tested: NF90-SW30 and XLE-SW30 where a gain in water quality is associated with a decrease in water productivity. We have to find the optimized parameters for each configurations.

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